

PATENT APPLICATION

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re the Application of

Hiroyuki MOCHIZUKI et al.

Group Art Unit: 1794

Application No.: 10/572,643

Examiner: B. CROUSE

Filed: March 20, 2006

Docket No.: 127380

For: ORGANIC ELECTROLUMINESCENT ELEMENT AND MANUFACTURING
METHOD THEREOF

DECLARATION UNDER 37 C.F.R. §1.132

I, Hiroyuki MOCHIZUKI, a citizen of Japan, hereby declare and state:

1. I received a doctoral degree from the Interdisciplinary Graduate School of Science and Engineering, Tokyo Institute of Technology in March 2001, where my doctoral thesis was in the field of organic electroluminescent devices.
2. I have been employed by the National Institute of Advanced Industrial Science and Technology (AIST) since 2001 and I have had a total of more than 8 years of work and research experience in polymer opto-electronic devices.
3. I am a named inventor in the above-captioned patent application.
4. I have a professional relationship, as an employee, with an assignee of the above-identified patent application. In the course of that professional relationship, I received compensation directly from the assignee for my work relating to polymer opto-electronic devices. I am not being compensated for my work in connection with this Declaration, other than my regular compensation as an employee.

5. I and/or those under my direct supervision and control have conducted the following tests:

I. Doping Of An Unsubstituted Π Conjugated Organic Compound With A Charge Transport Material

Example A: Method of manufacturing an organic electroluminescent element of claims 1-4 of the instant application (Example A corresponds to Example 1 of the instant application).

As shown in Fig. 2 of the present application, 100 mg of an electron transport compound 2-(4-biphenyl)-5-(4-tert-butylphenyl)-1,3,4-oxadiazole (PBD) was placed, as an organic compound 20 having a vapor pressure, inside one end of a glass tube 10 with one closed end (e.g., with an outside diameter of 15 mm and an inside diameter of 12 mm, for example). Then, a resin thin film 30 (1 mm thick, 8 mm wide, 40 mm long) of PPV formed on a glass substrate having an ITO electrode was inserted into the center of the tube. The open end of the glass tube 10 was then connected to an evacuation device 50 to evacuate the inside of the tube.

Subsequently, as shown in Fig. 3, the portion of the glass tube 10 close to the open end connected to the evacuation device 50 was melted using a glass tube sealing burner 60, thus sealing the organic compound 20 and the resin thin film 30 inside a sealed glass tube 11, as shown in Fig. 4. After sealing, the sealed glass tube 11 was placed in a thermostatic chamber 70 for one hour with the internal temperature maintained at 120°C, and was then slowly cooled to room temperature for over one hour. Following this gradual cooling, the glass tube 11 was cut, and the resin thin film 30, into which the organic compound 20 had penetrated and dispersed, was removed. The quantity of PBD that had penetrated into the PPV was 2.7 wt%.

Then, silver and magnesium were deposited together to laminate a negative electrode, thereby producing an electroluminescent element. This electroluminescent element emitted yellowish green light, and had a maximum luminance of 3000 cd/m² at 14V. The external quantum efficiency was 3.2 lm/w.

Comparative Example A: Method of manufacturing an organic electroluminescent element using a conventional spin coating method

When PPV and 2.7 wt% of PBD were placed in 100 ml of chloroform and stirred for 24 hours, the PBD dissolved in the chloroform, while the PPV remained completely insoluble. With this mixed liquid, film formation was impossible using inkjet methods, blade coating methods, or screen printing methods, meaning an electroluminescent element could not be produced.

Furthermore, 2.7 wt% of PBD was also mixed with a precursor of aqueous solution of PPV, but the PBD was also completely insoluble in this solution.

Comparative Example A': Method of manufacturing an organic electroluminescent element using a conventional contact and penetration method

PPV applied on a glass substrate cannot solvate in either water or organic solvent. On the other hand, although poly(p-xylene thiophenum chloride) as a precursor of PPV can solvate in water, PBD cannot solvate in the above poly(p-xylene thiophenum chloride) solution. Further, PBD cannot solvate in a solution in which poly(p-xylene thiophenum chloride) is solvated in methanol.

II. Doping Of A Substituted Π Conjugated Organic Polymer Compound With A Charge Transport Material

Comparative Example B: Method of manufacturing an organic electroluminescent element using the method of Example A.

As shown in Fig. 2 of the present application, 100 mg of an electron transport compound 2-(4-biphenyl)-5-(4-tert-butylphenyl)-1,3,4-oxadiazole (PBD) was placed, as an organic compound 20 having a vapor pressure, inside one end of a glass tube 10 with one closed end (and with an outside diameter of 15 mm and an inside diameter of 12 mm, for example). Then, a resin thin film 30 (1 mm thick, 8 mm wide, 40 mm long) of MEHPPV (poly(2-methoxy-5-(2'-ethyl-hexoxy)-1,4-para-phenylenevinylene)) formed on a glass substrate having an ITO electrode was placed inside the tube. The open end of the glass tube 10 was then connected to an evacuation device 50 to evacuate the inside of the tube.

Subsequently, as shown in Fig. 3, a portion of the glass tube 10, near the open end connected to the evacuating device 50, was melted using a glass tube sealing burner 60, thus sealing the organic compound 20 and the resin thin film 30 inside the sealed glass tube 11, as shown in Fig. 4. After sealing, the sealed glass tube 11 was placed in a thermostatic chamber 70, as shown in Fig. 5, was kept in the thermostatic chamber 70 for one hour with the internal temperature maintained at 120°C, and was then slowly cooled to room temperature over one hour. Following this gradual cooling, the glass tube 11 was cut, and the resin thin film 30, into which the organic compound 20 had penetrated and dispersed, was removed. The quantity of PBD that had penetrated into the MEHPPV was 2.7 wt%.

Then, silver and magnesium were deposited together to laminate a negative electrode, thereby producing an electroluminescent element. This electroluminescent element emitted vermillion colored light, and had a maximum luminance of 2500 cd/m² at 14V. The external quantum efficiency was 3.1 lm/w.

Comparative Example B': Manufacturing method using a conventional spin coating method.

MEHPPV and 2.7 wt% of PBD were added to chloroform, and following stirring, the solution was used to form films on glass substrates comprising ITO films. Visual inspection

showed that PBD crystalline portions existed within a number of the formed films, indicating that phase separation had occurred. Subsequently, silver and magnesium were deposited together to laminate a negative electrode, thereby producing an electroluminescent element. This electroluminescent element emitted vermilion-colored light, and had a maximum luminance of 900 cd/m^2 at 14V. The external quantum efficiency was 0.9 lm/w.

III. Discussion

6. In the development of a display monitor, the luminance must be approximately 1000 cd/m^2 or greater, although it may vary depending on the fineness of the pixels. If the luminance is less than 1000 cd/m^2 , it may not be possible to recognize an image in a typical room environment (under fluorescent light). If the efficiency is 1 lm/w or less, then power consumption is large, normal batteries are consumed in just several minutes of lighting, and the heating value is so high that the element itself may be damaged. The unit "lm/w" used herein denotes "lumen/watt."

7. From the results of Comparative Example B and Comparative Example B', it is evident that, even when manufacturing an organic electroluminescent element using a substituted π conjugated organic polymer compound such as MEHPPV, which displays better applicability to processing such as doping than unsubstituted π conjugated organic polymer compounds, an organic electroluminescent element with the required maximum luminance at 14V of at least 1000 cd/m^2 , and the required external quantum efficiency of at least 1 lm/w, cannot be produced unless the electron transport compound is converted to gas molecules.

8. The results of Example A and Comparative Examples A and A' show that when manufacturing an organic electroluminescent element using the unsubstituted π conjugated organic polymer compound PPV, the organic electroluminescent element cannot be produced unless, as described in the present application, the electron transport compound is converted to gas molecules.

9. I hereby declare that all statements made herein of my own knowledge are true, and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine and/or imprisonment under Section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the application or any patent issuing therefrom.

Date: April 13 2009

H. Mochizuki
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